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LONG-LIVED RADIOACTIVITY ASSOCIATED WITH RESPIRABLE QUARTZ DUST IN HARD ROCK UNDERGROUND URANIUM MINES

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LONG-LIVED RADIOACTIVITY ASSOCIATED WITH RESPIRABLE QUARTZ DUST IN HARD ROCK UNDERGROUND URANIUM MINES

by

J. Bigu*

ABSTRACT

The long-lived radioactivity associated with quartz dust samples taken in hard rock underground uranium mines has been measured. The total respirable dust was estimated by the mining company; respirable quartz dust was measured at the Elliot Lake Laboratory using X-ray diffraction techniques. The long-lived radioactivity in the samples was measured by conventional α-particle detection methods. The long-lived α-radioactivity was plotted against quartz dust content in the samples. A linear relationship was obtained which can be expressed by the equation $A = mW_Q + b$, where A and W_Q represent the α-activity (mBq) and quartz mass (mg) in the sample, respectively. The data also show that about 80% of the samples taken had long-lived radioactivity concentrations below 50 mBq/m^3, whereas approximately 90% of the samples selected had respirable quartz dust concentrations below 0.6 mg/m^3.

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RADIOACTIVITÉ À LONGUE PÉRIODE ASSOCIÉE AUX POUSSIÈRES DE QUARTZ INHALABLES DANS LES MINES D'URANIUM SOUTERRAINES DE ROCHE DURE

par

J. Bigu*

RÉSUMÉ

La radioactivité à longue période associée aux poussières de quartz inhalables contenus dans des échantillons prélevés dans les mines d'uranium souterraines de roche dure a été mesurée. Les poussières inhalables totales ont été estimées par la compagnie minière; les poussières de quartz inhalables ont été mesurées au Laboratoire d'Elliot Lake à l'aide des techniques de diffraction des rayons X. La radioactivité à longue période des échantillons a été mesurée à l'aide des méthodes classiques de détection des particules alpha. Le tracé de la radioactivité alpha à longue période par rapport à la poussière de quartz contenue dans les échantillons a été effectué. Un rapport linénaire a été obtenu, et peut être rendu par l'équation $A = mW_Q + b$, dans laquelle A et W_Q représentent respectivement l'activité alpha (mBq) et la masse du quartz (mg) de l'échantillon. De plus, les données indiquent qu'environ 80 % des échantillons prélevés contiennent des concentrations de radioactivité à longue période au-dessous de 50 mBq/m^3 alors qu'environ 90 % des échantillons choisis ont des concentrations de poussières de quartz inhalables au-dessous de 0,6 mg/m^3.

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INTRODUCTION

Inhalation of the short-lived decay products of radon is of great concern from the occupational health viewpoint in underground uranium mines, and other uranium-related industries. However, in the last few years increased interest has also been expressed about the potential health risks associated with the inhalation of the short-lived decay products of thoron, ^{238}U and ^{232}Th and their long-lived decay products.

^{238}U and ^{232}Th and their long-lived decay products are usually associated with airborne dust in the respirable size range (1 - 10 μm), as well as with dust of size greater than 10 μm. Long-lived radioisotopes associated with dust in the respirable size range, and beyond, are usually referred to as long-lived radioactive dust (LLRD). Concern about inhalation of LLRD is expressed because, once inhaled and lodged in tissue, they will remain active for very long periods, much longer than human life, unless eliminated by natural biological processes.

In hard rock uranium mines, long-lived radioactive dust is presumed to be associated with airborne quartz dust generated in a number of mining operations. In order to verify this contention, the long-lived radioactivity associated with dust samples collected in underground uranium mine atmospheres was measured. The activity measured was related to the dust quartz content in the samples.

EXPERIMENTAL PROCEDURE

Dust samples from a variety of hard rock mines and of mine working sites are routinely taken with specially designed gravimetric samplers (CAMPEDS) (1,2) and nylon cyclones, and sent to the Elliot Lake Laboratory for quartz analysis by X-ray diffraction techniques. For the purpose of the

present study, samples from two hard rock underground uranium mines, heretofore referred to as Mine A and Mine B, were supplied to the Elliot Lake Laboratory with the dust samples collected on silver membrane filters (5 μm pore size), as well as total dust mass, sampling flow-rate, volume of air sampled, and other relevant data.

Quartz dust was estimated by X-ray diffraction analysis. The long-lived radioactivity on the samples was measured by gross α-count using standard techniques. Radioisotope identification was carried out by α-spectrometry and γ-spectrometry (3,4).

Radioactivity measurements were made not earlier than one month after the dust samples were taken. This procedure was followed to ensure, quite conservatively, complete decay of airborne short-lived decay products, i.e., radon daughters and thoron daughters, collected during the dust sampling period.

A representative range of values for the quartz dust mass sampled in the filters was selected from the thousands of samples collected over the years. The total dust mass ranged from $\leq$0.02 mg to $\geq$0.4 mg. Samples taken over the period 1983-85 were selected for the study.

Gross α-activity in each sample was measured as follows. Three sequential 10-min background α-counts were taken followed by three sequential 30-min α-counts with the sample in place. The average values for the background and sample α-count, in counts per minute (cpm), were calculated, and the difference in α-count, i.e., net α-count, was computed.

Net α-count, in cpm, was converted into absolute disintegration rate, in disintegrations per minute (dpm). The disintegration rate was converted into activity in mBq. The formulae used are:

$$\text{cpm} = \left(\frac{\overline{N}_\alpha}{\overline{T}_\alpha} - \frac{\overline{B}}{\overline{T}_B} \right) \qquad \text{Eq 1}$$

$$dpm = cpm/\epsilon \qquad \text{Eq 2}$$

$$A(mBq) = 10^3(dpm/60) = 16.67\ dpm \qquad \text{Eq 3}$$

where, in the above expressions, $\bar{N}_\alpha$ and $\bar{B}$ stand for the average sample gross α-count, over time T_α, and average background α-count over the time T_B, respectively. The symbol ϵ represents the α-counting efficiency of the instrument used to measure gross α-count. The letter A represents the activity of the sample in mBq. It should be noted that by definition 1 Bq = 1 dps, where dps = dpm/60, i.e., disintegrations per second.

Equations 1, 2 and 3 can be written as one:

$$A(mBq) = \frac{16.67}{\epsilon}\left(\frac{\bar{N}_\alpha}{T_\alpha} - \frac{\bar{B}}{T_B}\right) \qquad \text{Eq 4}$$

The activity, A, was plotted against quartz mass, in mg. The specific activity concentration levels, A_S, were tabulated. A_S was calculated according to:

$$A_S = 10^3 A/QT_S \qquad \text{Eq 5}$$

where Q is the sampling flow-rate in L/min and T_S is the sampling time in min. As A is in mBq, A_S is given in mBq/m^3.

A total of 313 samples was analyzed, i.e., 76 samples (1983), 121 samples (1984), and 116 samples (1985). The number of samples analyzed per year was decided according to the available range of dust mass suitable for gross α-count measurements, and radioisotope identification by α- and γ-spectrometry. Samples were taken using the CAMPEDS and nylon cyclones.

Data were analyzed on a yearly basis for each mine. The three independent sets of data corresponding to 1983, 1984 and 1985, respectively, were compared to identify possible differences arising from changes that may have occurred in ore grade, mining operations, sampling locations and methods. Finally, the data collected over the 1983-85 period for each mine were analyzed as a single set of data.

Data were plotted as frequency histograms for quartz dust mass and long-lived radioactivity. In addition, long-lived radioactivity was plotted against quartz dust mass and total dust mass.

EXPERIMENTAL RESULTS AND DISCUSSION

Figures 1 to 4 show frequency histograms for respirable quartz dust (Figures 1 and 2), and long-lived radioactivity of dust samples (Figures 3 and 4). Figures 5 and 6 show the long-lived radioactivity associated with quartz dust content. Figures 1 to 4 represent data collected during the period 1983-85. Figures 1, 3 and 5,, and Figures 2, 4 and 6 correspond to Mine A and Mine B, respectively.

Figures 1 and 2 show frequency histograms of airborne quartz dust concentration (mg/m^3). The graphs indicate that high quartz dust concentrations are less frequent than low quartz dust concentrations. The sample percentage, of the total, corresponding to a given quartz dust concentration range is also indicated in both Figures. These data show that 80% to 90% of the samples selected correspond to quartz dust concentrations below 0.6 mg/m^3.

Figures 3 and 4 show frequency histograms of the airborne long-lived radioactivity concentration (mBq/m^3) associated with the quartz dust samples. Also shown in the graph is the sample percentage, of the total, corresponding to a given long-lived radioactivity range. The data show that about 80% of the samples taken have long-lived radioactivity concentrations below 50 mBq/m^3.

Figures 5 and 6 show the long-lived radioactivity associated with the dust samples, versus quartz dust mass for the period 1983 to 1985. Also shown are the best-fitted straight lines obtained by linear regression analysis. Figures 5 and 6 show that the data can be approximated by the following linear

relationships:

$$A(mBq) = 160.25\ W_Q + 7.87 \text{ (Mine A)} \qquad \text{Eq 6}$$

$$A(mBq) = 166.88\ W_Q + 2.22 \text{ (Mine B)} \qquad \text{Eq 7}$$

where W_Q represents the quartz dust mass in mg. The slopes and intercepts for the combined period 1983 to 1985 did not differ significantly, i.e., within the limits of experimental error, from the average values taken from data corresponding to 1983, 1984 and 1985, separately, for each mine. Furthermore, Equations 6 and 7 show that there is no significant difference between mines A and B.

As previously indicated, a similar analysis to that shown above for total respirable dust, instead of respirable quartz dust, shows a significantly lower correlation coefficient. It is, therefore, concluded that there is a reasonably good linear correlation between the long-lived radioactivity associated with airborne dust in the respirable size range, and the quartz dust content in the dust sample. No such good correlation exists, however, for total respirable dust. This is to be expected as the other major component of total respirable dust, in the present case, is non-mineral dust, e.g., diesel particulates generated by tools, machinery and vehicles used in underground mining operations.

It should be noted that the large data scatter shown in Figures 5 and 6 is mainly due to problems associated with the handling of the samples as dust can easily be shaken off during handling. Furthermore, self-absorption effects have been ignored in this analysis. Absorption of α-particles in dust, i.e., self-absorption, becomes increasingly important with increasing dust thickness.

Equations 6 and 7 suggest that the airborne long-lived radioactivity associated with respirable dust can be approximately estimated if the quartz content in the sample is known, and vice versa. However, the reader should be

aware that, strictly speaking, the above relationships apply only to specific cases, and mine locations, as the long-lived radioisotope content in the orebody and in mine air depend, respectively, on the mine location, and the mining operation. Another important consideration is the dust size distribution generated which is related to the mining operation.

Radioisotope identification was conducted by α- and γ-spectrometry using, respectively, a silicon-barrier detector and a high purity germanium detector. The results are identical to those reported previously elsewhere (3,4).

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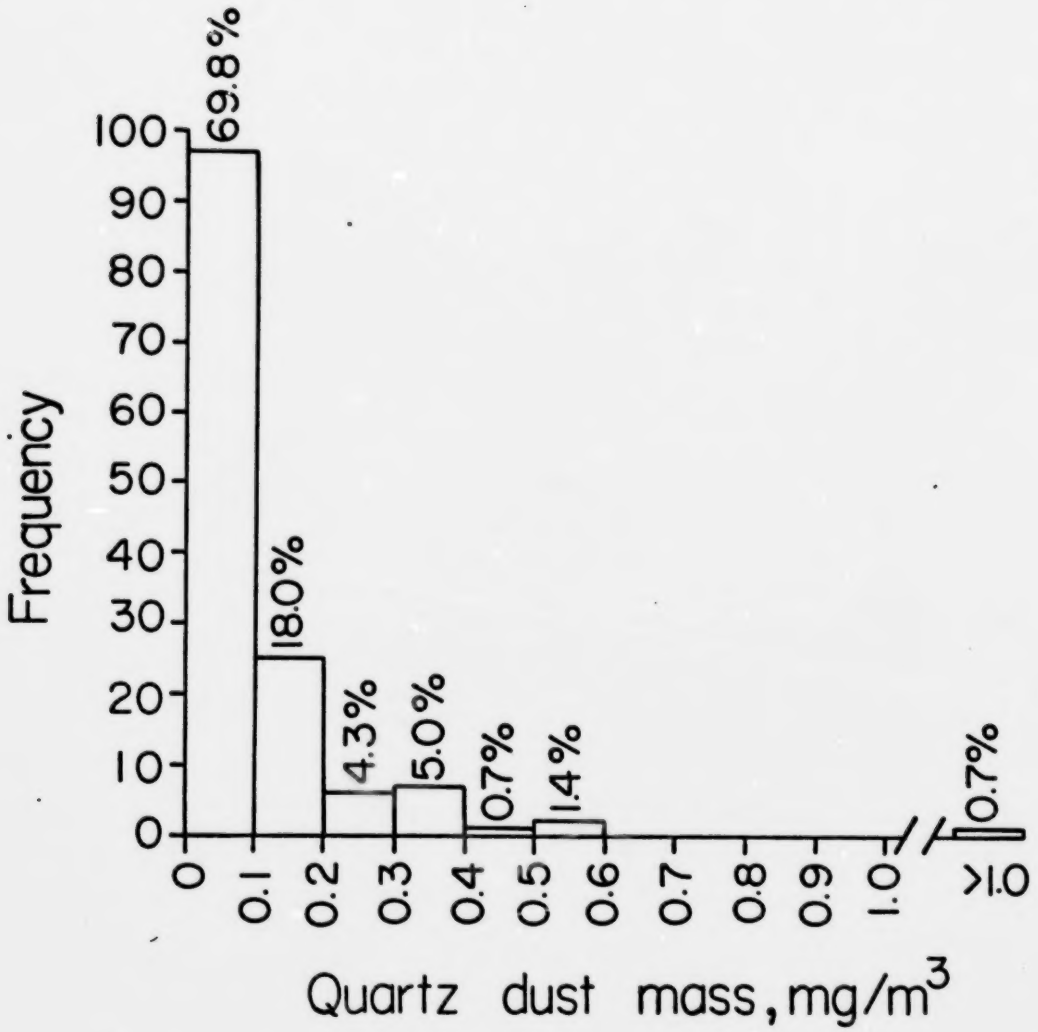

Fig. 1 - Frequency (distribution) versus quartz dust concentration. Also shown are the frequency percentages of the total number of measurements. (Mine A).

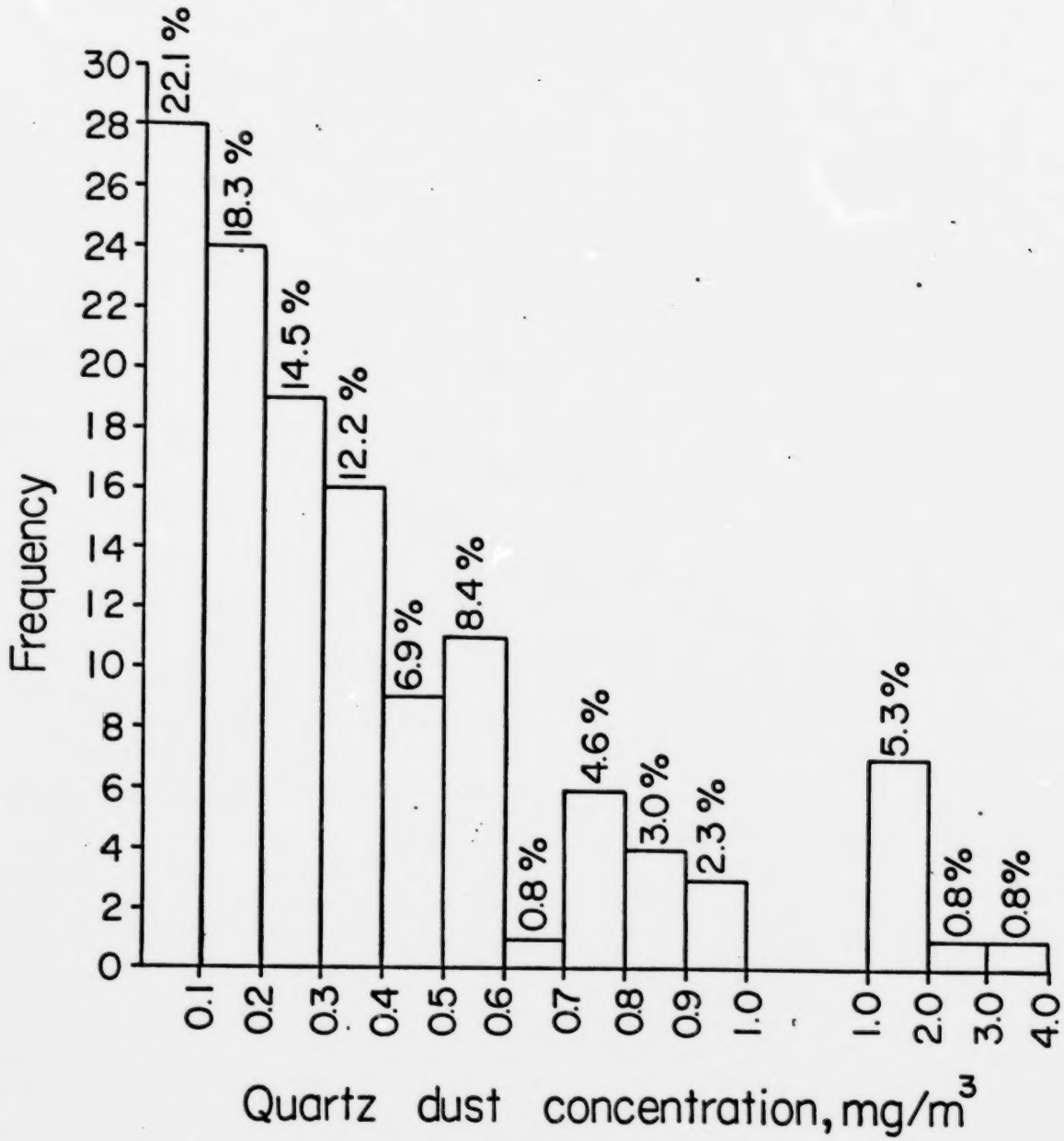

Fig. 2 - Frequency (distribution) versus quartz dust concentration. Also shown are the frequency percentages of the total number of measurements. (Mine B).

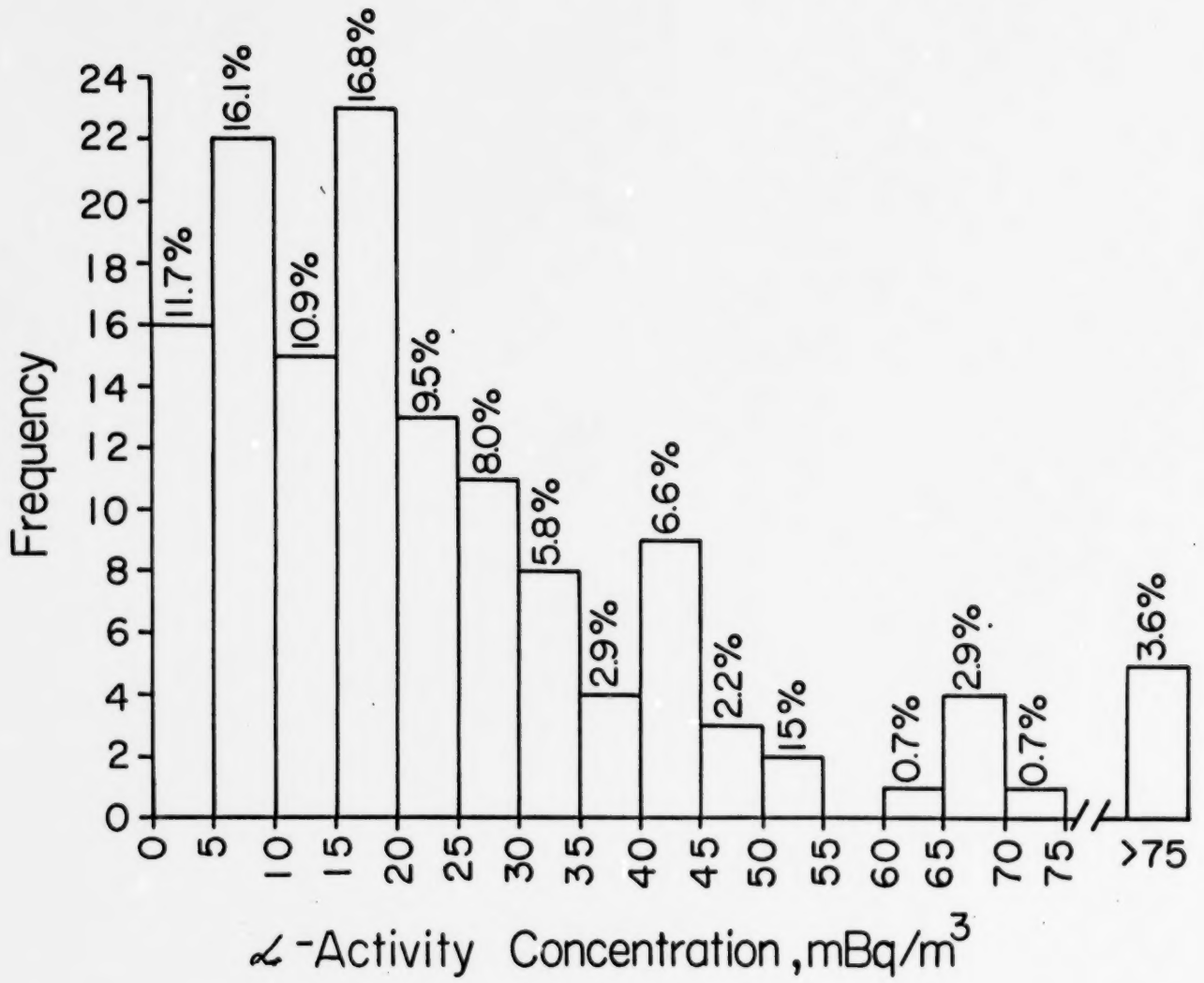

Fig. 3 - Frequency (distribution) versus α-particle activity concentration. Also shown are the frequency percentages of the total number of measurements. (Mine A).

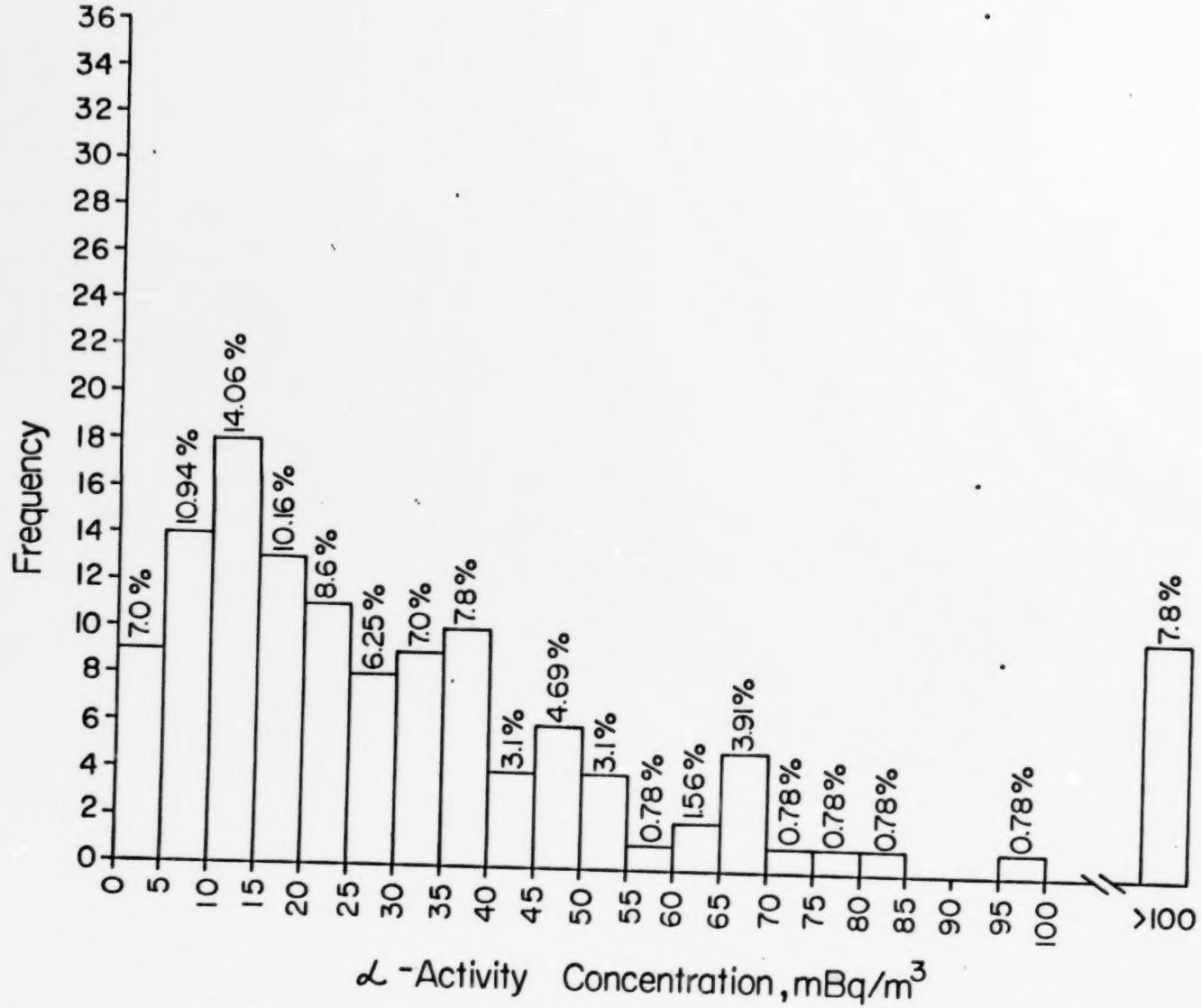

Fig. 4 - Frequency (distribution) versus α-particle activity concentration. Also shown are the frequency percentages of the total number of measurements. (Mine B).

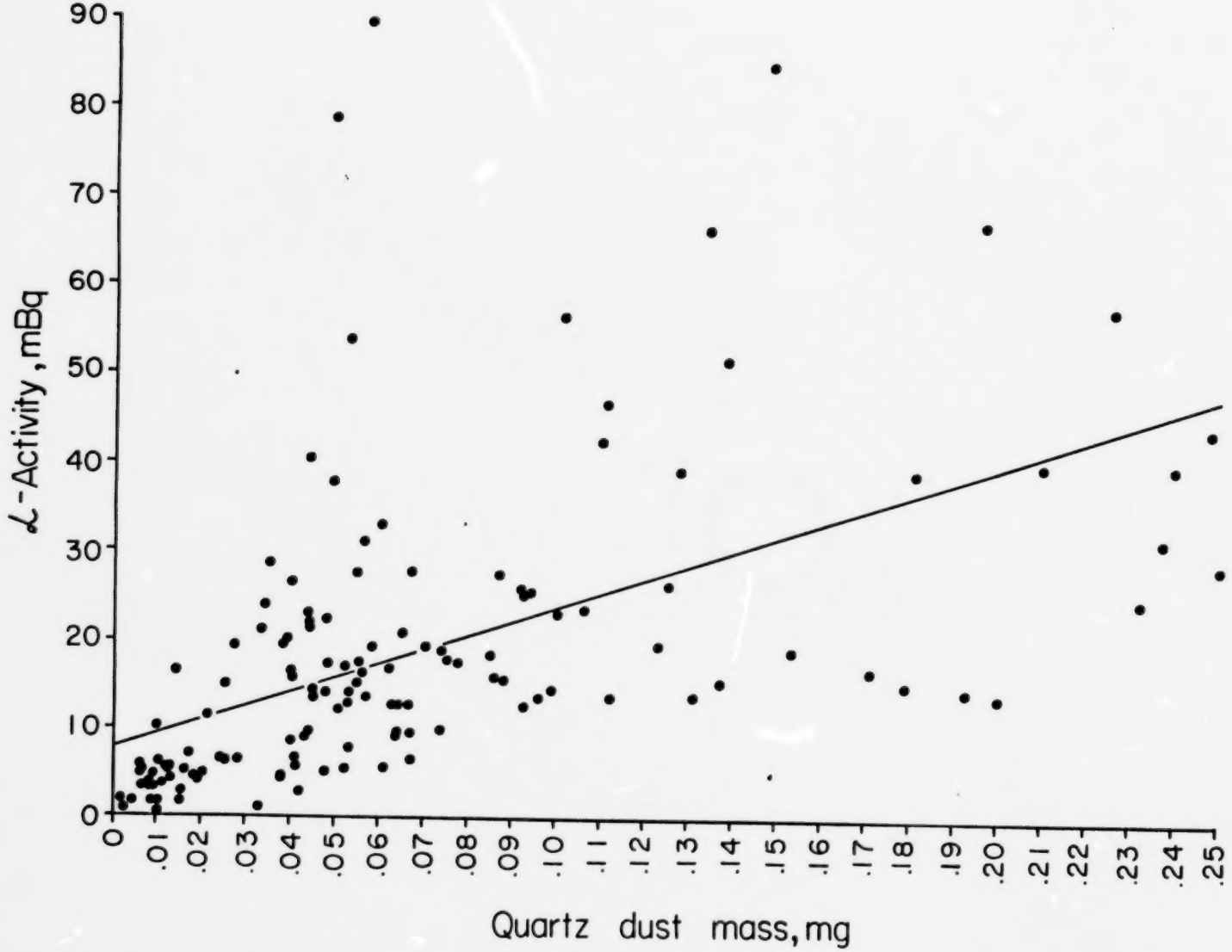

Fig. 5 - Alpha-particle activity versus quartz dust mass for the period 1983-1985. (Mine A).

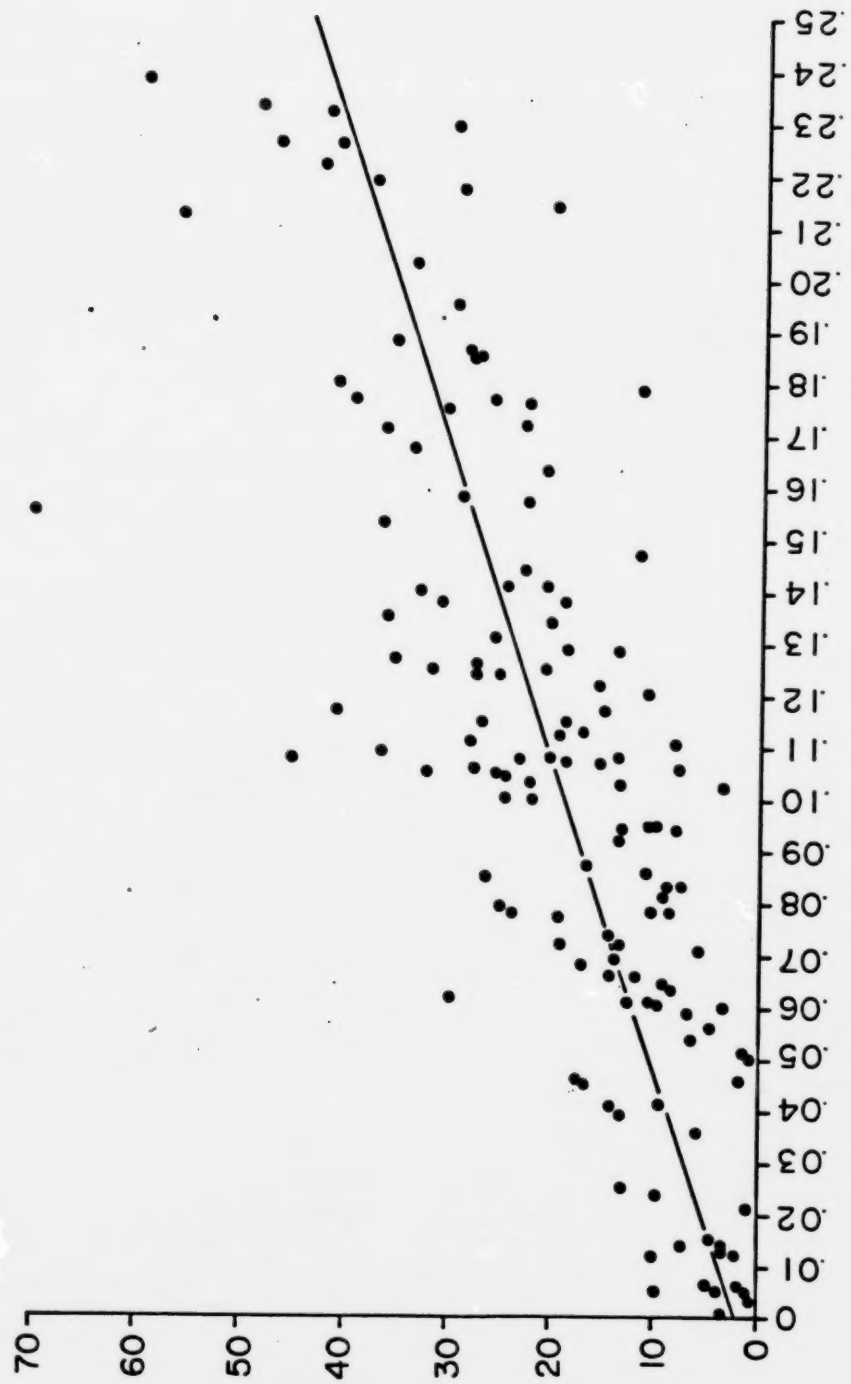

Fig. 6 - Alpha-particle activity versus quartz dust mass for the period 1983-85. (Mine B).